PREDICTING INDOOR PM2.5 OF OUTDOOR ORIGIN: TESTING A TRANSIENT SIZE-RESOLVED MODEL USING INTENSIVE MEASUREMENTS FROM A RESIDENCE

ML Fischer[1], MM Lunden[1], TL Thatcher[2], RG Sextro[2], and NJ Brown[1]

[1] Atmospheric Processes, Effects, and Analysis Program, Lawrence Berkeley National Laboratory, Berkeley, CA, USA
[2] Indoor Environment Dept., Lawrence Berkeley National Laboratory, Berkeley, CA, USA

ABSTRACT

We report tests of a model for indoor PM2.5 of outdoor origin that incorporates physical mechanisms for time dependent transport, and size dependent penetration and deposition. This work was performed using information obtained from an intensive study of a house near Fresno, CA, USA. During the multi-week study covering two seasons, we measured particles in both indoor and outdoor air, with high temporal, chemical, and size resolution, and other variables that also affect transport and fate. Results suggest that 1) the model captures a significant fraction of the variation in meteorologically forced air infiltration rate, 2) the predicted indoor/outdoor PM2.5 ratio is not consistent with the measured ratio unless a large (unphysical) deposition rate > 2 hr^{-1} is assumed, and 3) the differences between model and measurement in indoor PM2.5 are likely due to loss of volatile ammonium-nitrate aerosol. We conclude that nitrate particle volitization must be included in the model formulation.

INDEX TERMS

Measurements, Models, Outdoor, Particulate Matter, Ventilation

INTRODUCTION

Exposure to air containing particulate matter smaller than 2.5 microns in size (PM2.5) has been implicated in increased mortality and morbidity (Samet et al., 2000). Because personal exposures to outdoor sources of PM2.5 are better correlated with indoor than outdoor concentrationn, models have been developed to estimate indoor concentrations of outdoor origin (Thornburg et al., 2001; Riley et al., 2002). In particular, recent work by Riley et al. (2002, hereafter RMLN) incorporated current understanding of physical factors governing the indoor proportion of outdoor particles (IPOP), and the typical size distributions of outdoor particles, to estimate steady state probability distributions for IPOP and PM2.5 for urban and rural residences and small commercial buildings. This work showed that typically 0.3 < IPOP < 1, which is consistent with earlier work. Herein, we construct and test a transient physical model based on parameterizations from RMLN, and compare the results with measurements made at a residential research house.

METHODS

The building used for this study is an unoccupied single story (134 m^2 area) suburban residence in Clovis (a suburb of Fresno), CA. Measurements were made from August, 2000 to January, 2001, with three intensive measurement periods in October and December, 2000, and January, 2001. More information on the instrumentation and experimental conditions is provided in Lunden, et al. (2002a,b) and Thatcher et al. (2002a).

The model used to estimate indoor PM2.5 of outdoor origin is expressed as a 1st order time dependent linear differential equation for the concentration of indoor PM, $C_{in,j}$, in terms of outdoor concentration, $C_{out,j}$,

$$dC_{in,j}/dt = P_j(\Delta p,x)\, C_{out,j}\, \lambda - C_{in}\, (\, \beta_j(x) + \lambda), \text{ where} \tag{1}$$

the air infiltration rate into the building λ, the fraction of particles penetrating the building shell $P_j(\Delta p,x)$, and the particle deposition rate inside the building $\beta_j(x)$ are functions of particle size j, indoor/outdoor pressure difference, Δp, and other building factors, x, such as building furnishings and the turbulence intensity of indoor air. Particle penetration factor and deposition rate were estimated using the work of Liu and Nazaroff (2000), and Lai and Nazaroff (2001) and Thatcher et al. (2002b) respectively, under assumptions similar to those in RMLN. Curves showing typical ranges for size dependent penetration and deposition rate are shown in Figure 1. Taken together, these curves show that 0.1 to ~ 1 μm particles are expected to transport into a building and deposit at rates in a range (0.1 – 1 hr^{-1}) that is comparable to typical residential ventilation rates.

The air infiltration rate into the house is predicted for periods without active mechanical forcing using the LBNL/AIM infiltration model (Walker and Wilson, 1998), measured indoor-outdoor temperature differences, roof level wind speed and direction, and building leakage area as measured with the blower door method (Dickerhoff, 2001). The predicted infiltration rate is compared with the measured infiltration rate determined with a continuous SF_6 tracer gas measurement.

Indoor and outdoor particle number data were measured every 5 minutes with matched light scattering and aerodynamic particle sizing instruments, and binned into 20 logarithmically spaced bins from 0.1 to 2.5μm in size (Lunden, 2002b). Predicted indoor particle numbers are then calculated using the measured outdoor particle number data, the size dependent parameters from Figure 1, and the measured and predicted infiltration rates from Figure 2 as inputs to a forward marching finite difference form of Eq (1). Total PM2.5 concentration is calculated as the sum of size resolved particulate numbers assuming spherical particles with uniform density. Note that although uncertainty in the estimate of mass concentration is expected (due to uncertainties in the absolute instrument collection efficiency, particle shape and density), the estimated indoor/outdoor PM2.5 ratio is somewhat insensitive to these errors. Finally, concentrations are averaged over 12 hr intervals.

RESULTS

Measured and predicted ventilation rates from a period in December, 2000 to January, 2001, are shown in Figure 2. After active experimental manipulations of the house in late Dec., the air infiltration was dominated by thermal and wind induced indoor/outdoor pressure differentials (see inset of Figure 2). Linear regression of predicted on measured infiltration rates for this later period reveals that the predicted infiltration explains 64% of the variance in the measured data with a best fit slope of 0.65 +/- 0.02 and negligible offset. This suggests that the infiltration model captures most of the meteorologically driven variations but that a multiplicative uncertainty is present in some combination of the estimates of the model coefficients, building parameters, or measured air exchange rate. Below, we examine the effect of uncertainty in infiltration rate on the predicted indoor/outdoor PM2.5 ratio.

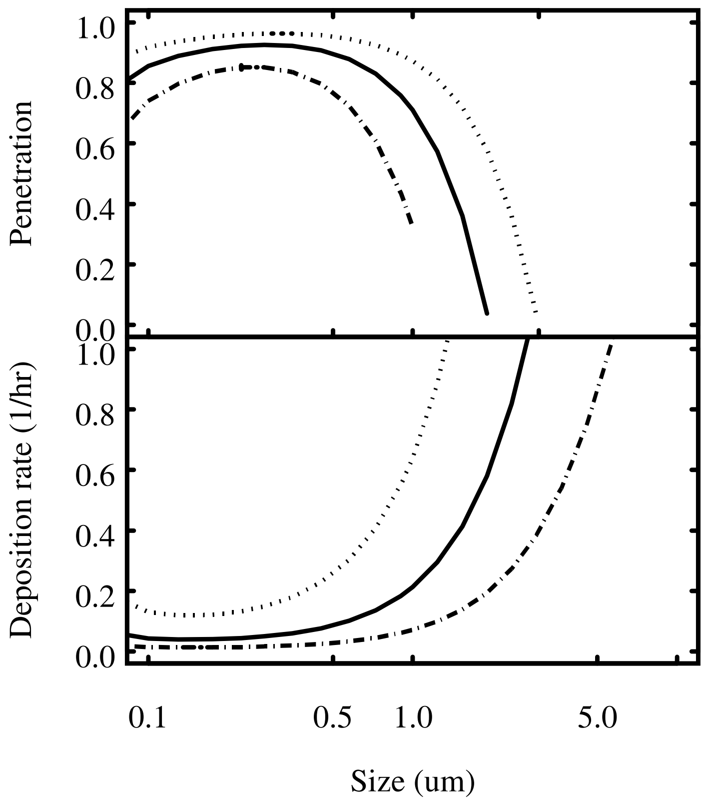

Figure 1. Size dependent particle penetration and depositions rates. Penetration is shown for indoor/outdoor pressures differences of 1 Pa (dotted), 0.3 Pa (solid), and 0.1 Pa (dashed). Deposition rate is shown as a mean estimate (solid) within a factor of 3 variations indicative of range of estimates from literature.

The measured and predicted ratios of indoor to outdoor PM2.5 are shown in Figure 3. In contrast to expectations, the values were measured to be significantly less than unity except during periods when outdoor air was rapidly forced into the building. The low ratio can, however, be explained if a very high (> 2 hr^{-1}) deposition rate is adopted. Additionally, the lower panel of Figure 3 shows that the uncertainty introduced by the predicted versus measured infiltration rate is relatively small. Similarly, using the upper estimate of deposition rates from Figure 1 does not have a very strong influence on the indoor/outdoor ratio. These two results can be explained by examining the steady state solution to Eq (1), $C_{in,j}$ (steady state) = $C_{out,j}\ \lambda/(\lambda+\beta)$. When infiltration and deposition are approximately equal variations in either parameter have a less than linear effect on the indoor/outdoor ratio.

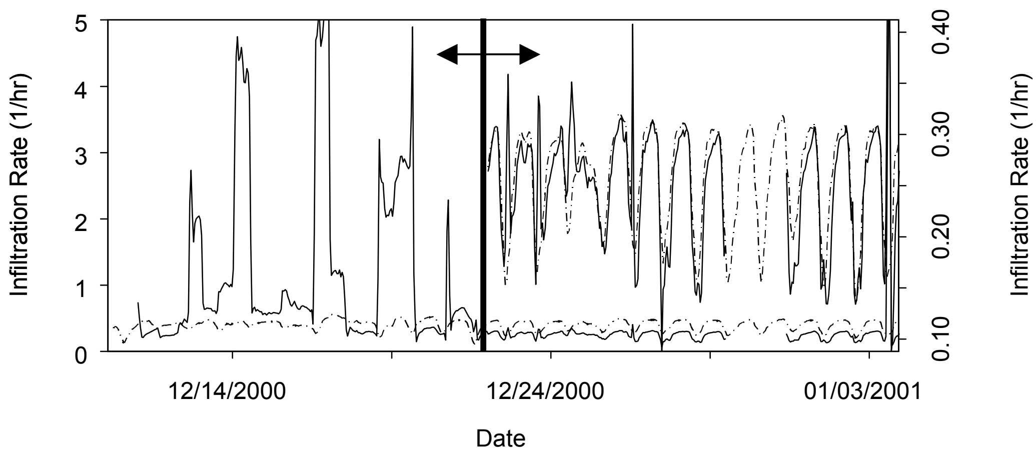

Figure 2. Measured (solid line) and predicted (dashed line) air exchange rates for period of winter measurements. To show the correlation between measured and predicted infiltration rate when building was not mechanically forced, the right portion of the graph is magnified (see right hand scale) and shows the measured and predicted infiltration rates with the predicted values scaled by a factor of 0.65.

DISCUSSION

The principal result of this work is that the measured indoor/outdoor PM2.5 ratio was significantly lower than can be explained by current information on the transport and loss of non-volatile aerosol species. This is likely due to the prevalence of volatile ammonium nitrate aerosol that vaporizes upon entry into the house (Lunden et al. (2002a)). The results obtained here are also consistent with those of Thatcher et al. (2002a), where indoor/outdoor differences in sulfate aerosols are well predicted using the physically based model of Eq (1) to obtain best fit penetration and deposition rates in the range given in Figure 1, while a high deposition rate was necessary to fit the measured indoor nitrate aerosol. Where volatile species constitute a significant fraction of total fine aerosol, successful predictions of indoor PM2.5 require a model that includes gas-particle partitioning.

These results also suggest that natural infiltration rates can be predicted with sufficient accuracy to allow prediction of non-volatile indoor PM2.5 ~ 10-20% precision. To produce a model for use in occupied buildings, further work capturing the effects of human occupancy on ventilation, penetration, deposition, and filtration may be required. Then, knowing regional characterizations of building leakage area, HVAC systems, and building operation, we expect that accurate estimates of the regional distribution of non-volatile indoor PM2.5 of outdoor origin will be possible. Inclusion of descriptions for these processes is underway.

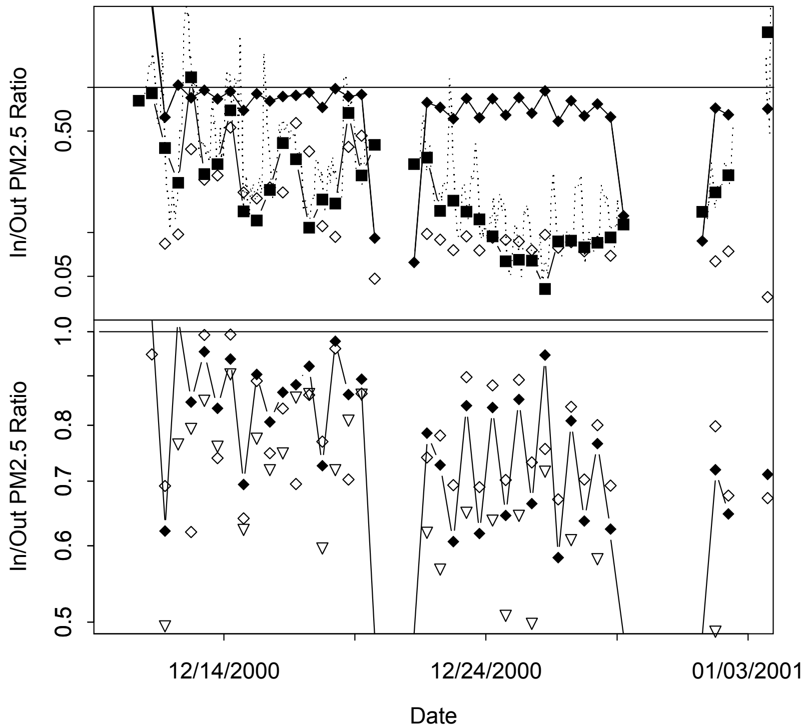

Figure 3. Indoor/outdoor PM2.5 ratio calculated from size resolved data and predicted by transient model. Upper panel compares measured ratio (dashed line, closed squares) against predicted ratio obtained with mid range model parameters and measured air exchange rate (solid line, closed diamonds) and predicted ratio obtained with deposition rate of 2.5 hr^{-1} (open diamonds). Note that a high deposition rate is necessary to approximately match the measured ratio. Lower panel (with expanded scale) shows predicted ratios obtained with mid range model parameters and measured infiltration rate (solid line, closed diamonds), mid-range model parameters and predicted infiltration rate (open diamonds), and upper estimate of deposition rate and measured infiltration rate (inverted triangles). Note that neither the over estimate of predicted infiltration rate nor the upper estimate of deposition rate produce more that 10-30% differences in predicted ratio.

CONCLUSION AND IMPLICATIONS

Measurements of air infiltration rate and size resolved fine particulate matter collected at a residence in the San Joaquin Valley, CA are compared with predictions from a building infiltration model and physically based transport model for non-volatile aerosol species. The results are considered in the context of producing a predictive model of indoor PM2.5 of outdoor origin that can be used at a regional level. The measured and predicted infiltration rates compare sufficiently well that the infiltration model can likely be used as part of an aerosol model. The aerosol transport model, however, does not capture the small indoor/outdoor PM2.5 ratios measured in the field. The reason for the discrepancy appears to be due to loss of volatile ammonium nitrate aerosol. Successful prediction of indoor PM2.5 in regions with significant concentrations of volatile aerosol, hence appears to require inclusion of physiochemical processes governing gas-particle phase changes. Analysis and inclusion of the appropriate descriptions for these processes is currently being conducted.

ACKNOWLEDGEMENTS
The authors gratefully acknowledge the advice and assistance of Dr. William Riley, Dr. Iain Walker, and Darryl Dickerhoff. This research was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Petroleum Technology Office under U.S. Department of Energy Contract No. DE-AC03-76SF00098 and by the Western States Petroleum Association.

REFERENCES
Dickerhoff DJ. 2001. (private communication)

Lai ACK, and Nazaroff WW. 2000. Modeling indoor particle deposition from turbulent flow onto smooth surfaces. *J. Aerosol Sci.* Vol 31, pp. 463-476.

Liu D, and Nazaroff WW. Modeling pollutant penetration across building envelopes. *Atmos. Environ.* Vol. 35, pp. 4451-4462.

Lunden MM, Thatcher TL, Littlejohn D, et.al. 2002a. The Transformation of Outdoor Ammonium Nitrate Aerosols in the Indoor Environment. Indoor Air 2002, *Proceedings of the 9th International Conference on Indoor Air Quality and Climate – Indoor Air 2002,* Vol 1. Santa Cruz: Indoor Air 2002.

Lunden MM, Thatcher TL, Littlejohn D, et.al. 2002b. Building a predictive model of indoor concentrations of outdoor PM-2.5 homes. Report Number: LBNL-48929. Lawrence Berkeley National Laboratory, Berkeley, CA, USA. URL: http://www-library.lbl.gov/docs/LBNL/489/29/PDF/LBNL-48929.pdf

Riley WW, McKone TE, Lai ACK, et al. 2002. Indoor particulate matter of outdoor origin: Importance of size-dependent removal mechanisms. *Environ. Sci. Technol*. Vol. 36, pp. 200-207.

Samet JM, Zeger SL, Dominici F, Curriero F, et al. 2000. The National Morbidity, Mortality and Air Pollution Study, Part II: Morbidity and Mortality from Air Pollution in the United States. Research Report 94, PartII. Health Effects Institute, Cambridge MA.

Thatcher TL, Lunden MM, Sextro RG, et.al. 2002a. The effect of penetration factor, deposition, and environmental factors on the concentration of PM2.5 sulfate, nitrate and carbon indoors. Indoor Air 2002, *Proceedings of the 9th International Conference on Indoor Air Quality and Climate – Indoor Air 2002,* Vol 1. Santa Cruz: Indoor Air 2002.

Thatcher TL, Lai ACK, Moreno-Jackson R, et al. 2002b. Effects of Room Furnishings and Air Speed on Particle Deposition Rates Indoors. *Atmospheric Environment* (in press).

Thornburg J, Ensor DS, Rodes CE, et al. 2001. Penetration of particles into buildings and associated physical factors. Part I: Model development and computer simulations. *Aerosol Science and Tech*. Vol. 34 (3), pp. 284-296.

Walker IS, and Wilson DJ. 1998. Field Validation of Algebraic Equations for Stack and Wind Driven Air Infiltration Calculations. *HVAC&R Research.* Vol. 4 (2), pp. 119-139.